LA-UR-17-28566
Approved for public release; distribution is unlimited.

Title: Demonstrations of Neutron Assay Systems

Author(s): Geist, William H.

Intended for: Training Course

Issued: 2017-09-21

DEMONSTRATIONS OF NEUTRON ASSAY SYSTEMS

Purpose

To provide participants with a demonstration on how various neutron NDA assay systems are used to assay nuclear material in the field.

Overview

Participants will perform mass measurements on nuclear material using the HLNC, AWCC, and UNCL. They will observe plutonium mass measurements using the HLNC. The AWCC will be used to measure the ^{235}U in bulk uranium items. The UNLC will be used to measure the ^{235}U linear mass in fuel assemblies.

Reference Documents

- PANDA Manual – Chapters 17
- LANL Report, *Description and Performance Characteristics for the Neutron Coincidence Collar for the Verification of Reactor Fuel Assemblies*, LA-8939-MS
- LANL Report, *Neutron Collar Calibration and Evaluation for Assay of LWR Fuel Assemblies Containing Burnable Neutron Absorbers*, LA-11965-MS

Contents

1. HLNC

a. Introduction

In this session you will use the High Level Neutron Coincidence Counter (HLNC) to assay plutonium items.

The principles of neutron coincidence counting are described in chapter 16 of the PANDA manual and the HLNC is described in chapter 17 of the PANDA manual.

The following three quantities are used frequently throughout this session:

1. Effective ^{240}Pu mass: the mass of ^{240}Pu that would produce the same coincidence rate from spontaneous fissions as the actual item; it is calculated from

$$\text{effective } {}^{240}\text{Pu} = 2.52\ {}^{238}\text{Pu} + {}^{240}\text{Pu} + 1.68\ {}^{242}\text{Pu},$$

where the Pu quantities are masses.

2. Multiplication: the number of neutrons leaving an item divided by the number of neutrons from spontaneous fissions and (alpha, n) reactions.

3. Alpha: the ratio of (alpha, n) neutrons to spontaneous fission neutrons.

The general idea in conventional passive neutron assay of plutonium items is to determine the effective ^{240}Pu mass from the doubles and singles rates and then to determine the Pu mass from the isotopic composition:

$$\text{Pu} = (\text{effective } {}^{240}\text{Pu}) \,/\, (2.52\, f_{238} + f_{240} + 1.68\, f_{242}),$$

where f_i is the weight fraction of isotope i.

The main problem with this is that the coincidence rate depends on the effective ^{240}Pu mass, the multiplication, and the alpha value; this is because induced fissions produce coincidence counts. Thus, in general, there are three unknown quantities: the effective ^{240}Pu mass, the multiplication, and alpha. In conventional coincidence counting, only two quantities are measured (the singles and doubles rates), so the multiplication or alpha value must be known or assumed to determine the Pu mass.

You will use the HLNC to assay Pu in two ways: by calibration curve and by known alpha multiplication correction.

The calibration curve method uses a calibration of coincidence rate vs. effective ^{240}Pu mass. This method works well if the unknown items have the same characteristics as the calibration standards. Only one measured quantity is used (the doubles rate); the multiplication and alpha value are assumed to match the standards that were used to construct the calibration curve.

The known alpha, multiplication correction method uses the ratio of the doubles rate to the singles rate to determine the neutron multiplication and then corrects the doubles rate for multiplication. This method is also called the Ensslin-Boehnel multiplication correction technique or the two parameter analysis method. This analysis technique works well for pure materials for which the multiplication varies because of density or shape variations. For example, for pure Pu metal items, this method works much better than the calibration curve method. Because this method uses two measured quantities to determine the multiplication and the Pu mass, the alpha value must be known. Alpha is 0 for pure Pu metal and can be calculated for pure Pu oxide.

There are at least three other ways to perform assays with conventional coincidence counting:

1. Known multiplication: Here it is assumed that alpha is unknown, but that the multiplication is determined by the item mass and geometry.

2. Self-interrogation: Here it is assumed that the spontaneous fission neutrons are much fewer than the (alpha, n) neutrons.

3. Add-a-source: Here a ^{252}Cf source is temporarily added to the measurement cavity to determine a correction for the effect of moderation in the item (developed for drum measurements).

These three techniques will not be covered in this course.

b. Background Measurement

In this section you will measure the room background singles and doubles rates that will be subtracted from all the measurements that follow.

Remove any sources in or nearby the HLNC to at least 3 m away.
Select "Acquire | Background."
Enter 15 cycles of 20 s each.
Record your results in the table below.

Background singles rate (1/s)	
Background doubles rate (1/s)	±

Usually the doubles rate should be less than 1 count/s. If it is not, there may be an electronics problem with your system.

c. Normalization Measurement

The purpose of this section is to perform a normalization measurement to be certain that the detector is still performing as it did at the time of calibration. There is little chance of a problem developing over a few hours in time, but normalization tests are very important for checking instrument performance over periods of months or years or after calibration.

Mount your reference ^{252}Cf source on the standard source rod and insert it into the hole in the top plug of the HLNC.

Select "Acquire | Normalization" and measure the source for 15 x 20 s.

Record your results in the table below.

Source id	
Doubles rate (1/s)	±
Expected/measured doubles rate	±
Pass/Fail	

The normalization test will fail if the new normalization constant differs from 1 by more than 3 standard deviations and by more than 4%.

d. Verification Measurement

In this section you will use your calibration curves to assay an item with the same item type that was used for the calibration measurements.

Place a pure plutonium item in the HLNC in the usual position—centered radially and raised 10 cm above the bottom plug.

Select “Acquire | Verification” and enter the following data:

Measure the item for 20 x 30 s and record your results in the tables below.

Item ID =		
Quantity	Value: Calibration curve method	Value: Known alpha method
Doubles rate (1/s)	±	±
Multiplication	******************	
Alpha	******************	
Assay Pu mass (g)	±	±
Declared Pu mass (g)		
Declared - assay mass (g)	±	±
% difference [(D-A)/D•100]	±	±
standard deviations difference		

Because your assay item is the same item type as your calibration curve standards, your assay mass should agree with the declared mass within 3 standard deviations.

2. AWCC

a. Introduction

In this session you will use the Active Well Coincidence Counter (AWCC) to assay enriched uranium metal or oxide items in fast mode.

The AWCC is described in chapter 17 of the PANDA manual.

Because the spontaneous fission rate of the uranium isotopes is very low, the AWCC uses americium-lithium (AmLi) neutron sources to induce fissions in the uranium. The fission neutrons are then counted with coincidence circuitry.

The induced-fission cross section for ^{235}U is much higher at low neutron energies than at high neutron energies, so the AmLi neutrons are moderated by polyethylene to reduce their energies and increase the fission rate.

There are two modes of operation for the AWCC: fast and thermal. In the fast mode, the sample cavity is lined with cadmium to remove the thermal neutrons; in the thermal mode, the cadmium is removed.

The fast mode is better suited to high-mass uranium items and the thermal mode is better suited to low-mass items. In the thermal mode, the high fission cross section improves the statistical precision of the measurement; this is useful for low-mass items. However, for high-mass items the high fission cross section results in poor item penetration; thermal neutrons induce most of the fissions and these occur near the surface of the item.

You will use a calibration curve for uranium metal or uranium oxide in the fast mode and perform a verification assays based on this calibration curve. Fast and thermal modes have much different calibration curves. Also, different kinds of materials have different calibration curves because the penetration of the AmLi neutrons into the items and the neutron multiplication by the items are both sensitive to the density and geometry of the items.

b. Normalization

The purpose of this section is to check the stability of the AWCC by comparing the singles rate from the AmLi sources measured before the calibration to that measured now.

Position the AmLi sources in the counter. Be sure to select the correct source and insert them in the proper orientation.

Remove any uranium items from the AWCC.

Select "Acquire | Normalization."

Measure the AmLi sources for 15 x 10 s.

Record your results:

AmLi expected singles rate	±
AmLi measured singles rate	±
Ratio (expected/measured)	±
New normalization constant	±

The new normalization constant is set to 1 if the calculated normalization constant is within 4% of 1. This 4% limit is the default value set in the INCC software. If your new normalization constant is not 1, typically the room background would be remeasured and the normalization measurement repeated.

c. Verification

In this section you will use your fast-mode calibration curve to perform an assay of an uranium item.

Place the item in the AWCC cavity centered on the 5 cm stand similar to what was used for the calibration. Select “Acquire | Verification” and enter the following data:

Item id	
Declared ^{235}U mass	
Count time (s)	30
Mode	“Use number of cycles”
Number cycles	20

Measure the item for 20 x 30s and record your results in the table below.

Quantity	Value
Doubles rate (1/s)	±
Assay ^{235}U mass (g)	±
Declared ^{235}U mass (g)	
Declared - assay mass (g)	±

Because your assay item is the same item type as your calibration standards, your assay mass should agree with the declared mass within three standard deviations.

3. UNCL

a. Introduction

The goal of this exercise is to use the UNCL system to verify the ^{235}U loading of a fresh BWR fuel assembly mockup. An active measurement technique will be used where random neutrons from an AmLi (α,n) source induce fissions in the ^{235}U contained within the assembly. The induced fissions are characterized by the emission of time-correlated neutrons and are counted using the same coincidence circuitry as the High Level Neutron Coincidence Counter (HLNC).

The system has four slabs of polyethylene surrounding the assembly: three contain ^{3}He gas proportional neutron detectors and the fourth contains the AmLi interrogation source. The basic references for the UNCL is entitled "Description and Performance Characteristics for the Neutron Coincidence Collar for the Verification of Reactor Fuel Assemblies," Los Alamos National Laboratory report LA-8939-MS and a more recent report, "Neutron Collar Calibration and Evaluation for Assay of LWR Fuel Assemblies Containing Burnable Neutron Absorbers," Los Alamos National Laboratory report LA-11965-MS (ISPO-323). Refer to these reports for the most up-to-date and comprehensive report on the UNCL.

A simulated BWR fuel assembly containing 36 rods enriched to 2.34% ^{235}U will be used for measurements. The linear loading of the assembly is 8.79 g ^{235}U/cm and the length of the fuel zone is 121.9 cm. The assembly will be measured full and if time permits with several rods removed to determine the response to rod removal or substitution of dummy rods. The neutron collar is also used to verify the fissile content of fresh PWR and WWER assemblies.

The collar measures ^{235}U in the region between the slabs; therefore, its response is proportional to the quantity of ^{235}U per unit length of assembly. The collars use by a safeguards inspector requires a previous calibration on fuel assemblies of known composition and enrichment. The inspector checks that the assemblies to be tested do not differ significantly from the expected response. If the AmLi interrogation source is removed, the detector measures time-correlated neutrons from ^{238}U spontaneous fission decay. This passive measurement is an additional verification tool for the inspector.

Calibration requirements and constraints are significantly different for inspector-based NDA equipment than for in-plant operator equipment. The traditional calibration approach of making up standards typical of the unknowns and measuring them on the same detector that will be used for the unknowns is impractical for field verification in several facilities around the world at once. Calibration and normalization procedures, which reduce the number of physical standards and the time required for calibration by an inspector, have been developed for the neutron collar.

The basic idea is to carefully calibrate <u>one</u> member of the UNCL family for an important category of material [for example, boiling-water-reactor (BWR) fuel assemblies] covering a wide range of mass loadings. The calibration parameters for this <u>reference</u> <u>detector</u> are then <u>fixed</u>, and the response from any other neutron collar is normalized to the fixed calibration parameters for verification measurements.

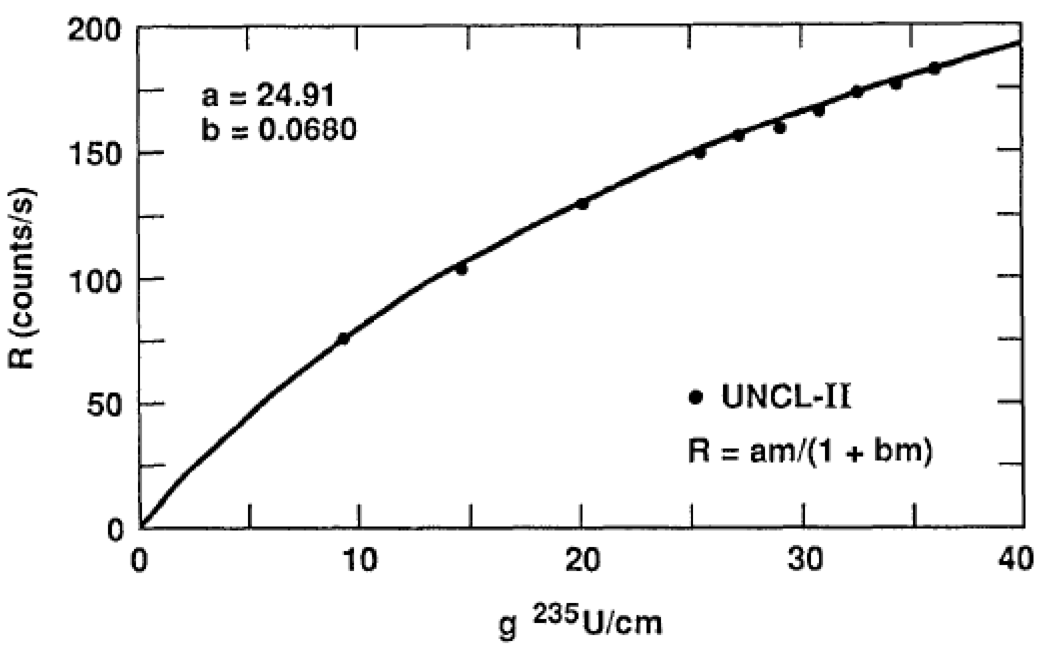
This normalization can be performed with a single fuel assembly characteristic of the material category. It is only necessary to count the same cross-reference fuel assembly in both the reference detector and the field detector, and to measure a reference source rate at the same time. This reference source (for example, ^{252}Cf or ^{241}AmLi) goes with the UNCL for field normalization.

This normalization can be performed with a single fuel assembly characteristic of the material category. It is only necessary to count the same cross-reference fuel assembly in both the reference detector and the field detector, and to measure a reference source rate at the same time. This reference source (for example, ^{252}Cf or ^{241}AmLi) goes with the UNCL for field normalization.

This technique of fixing the calibration parameters (curve shape) assumes that the nonlinear shape is mainly a property of the assemblies, and any detector-related effects are the same for all members of the UNCL family. The basic calibration function used with the UNCL is

$$R = \frac{aM}{1 + bM}$$

where M is the mass per unit length (g ^{235}U/cm) and a and b are calibration constants. This function is valid for LWR assemblies over the enrichment range from 1% to 4% ^{235}U. Figure 1 shows a typical calibration curve for BWR fuel assemblies (No Cd).



-4) collar

Because not all fuel assemblies and collars are identical, correction factors k_1 and k_2 must be defined to adjust the calibration function to different detector heads, AmLi sources, electronics units, burnable poisons, and assembly sizes. When these variables differ from our calibration condition, it is necessary to correct the measured response by

$$kR = (k0\times k1\times k2\times k3\times k4 \times k5)\,R$$

where

- AmLi source strength – k0
- Electronics drift – k1
- Detector efficiency and source yield – k2
- Burnable poison, Gd2O3 – k3
- Heavy metal loading, g U/cm – k4
- Other conditions – k5

These components of the correction factor are discussed below.

A. Electronics Normalization k1

Detector and electronic counting efficiency depends on the ^{3}He tubes and polyethylene moderator, the assembly-detector solid angle, and the electronics. The first of these is nearly constant for a given UNCL system. The second can be made essentially constant by very careful positioning of fuel assemblies within the collar. The electronics may change from time to time; however, if the high-voltage and discriminator settings are not altered, we usually obtain 1-2% stability in the totals rate.

The AmLi source used with the UNCL for fuel assembly verification is used also for electronics normalization. At the time of "cross-reference calibration" of the field collar with the "primary calibration" collar, the net totals rate (T_0) from the source was measured for the field detector. Then at the time of field use, the net totals rate (T_{field}) is remeasured; and any changes in field detector efficiency can be corrected for by the ratio

$$k(\text{elec}) = \left(\frac{T_0 \; e^{-\lambda \Delta t}}{T_{field}} \right)^2 \quad .$$

The ratio is squared because the totals rate varies with efficiency (ε), but the reals rate, R, varies as $(\varepsilon)^2$. Americium-241 has a decay half-life of 432 years; thus, if the time interval between T_0 and T_{field} measurements exceeds approximately 2 years, the decay correction, $e^{-\Delta\lambda\tau}$, must be made, where $\lambda = 0.0016 \text{ yr}^{-1}$ and Δt is the time in years since the calibration date.

If a different AmLi source is used for T_{field} than was used for T_0, an additional adjustment to T_0 is required before calculating k(elec) by the above equation, that is,

$$T_0(\text{corr}) = \frac{\text{Yield of AmLi field source}}{\text{Yield of AmLi calibraton source}} \quad .$$

B. Interrogation Source Decay k0

Because AmLi neutron sources decay, the coincidence response for even the reference collar with standard fuel assemblies would with time fall below the calibration curve. Therefore, to use the calibration parameters shown in Fig. 2, changes in coincidence response must be corrected for interrogation source decay by the factor

$$k(\text{source decay}) = e^{\lambda\Delta\tau} \quad .$$

C. Detector Efficiency and Source Yield k2

Different collars, even when fabricated by the same supplier, are likely to have small differences in coincidence response. Slight differences in the size of the polyethylene interrogation cavity and the location of the source hole will change the average neutron flux in the assembly per source neutron. Variations in polyethylene density may cause small differences in moderation. Also, not all ^{3}He tubes will demonstrate identical sensitivities.

A source-assembly-detector coupling correction can be determined by counting the same fuel assembly (or mockup) with <u>both</u> the reference (primary calibration) collar and the field collar, each with their respective AmLi sources. Then

$$k(\text{coupling}) = \frac{R_0\,(\text{reference collar, reference AmLi source})}{R_0\,(\text{field collar, field AmLi source})} \quad .$$

Note that in this form the factor corrects not only for differences in source-assembly-detector coupling but also for differences in the AmLi sources used in the field collar and the reference collar. Measured parameters for cross-reference calibration of a number of neutron collar units can be found in. LA-11965-MS (ISPO-323)

D. Burnable Neutron Absorbers k3

Most BWR assemblies contain burnable poison (Gd_2O_3) rods to provide longer fuel assembly lifetimes. The presence of strong neutron absorbers within the fuel assemblies affects the interrogation flux and thus the UNCL response.

The correction for neutron-absorber rods in BWR assemblies is made as follows:

$$k3\ (No\ Cd) = 1 + n\left(\frac{76}{N}\right)(corr\ per\ rod) \times\ \delta$$

where n is the number of absorber rods and the (corr per rod) is

$$(Corr\ per\ rod) = 0.0572(1 - e^{-0.647Gd})$$

and δ is an enrichment correction given by

$$\delta = (1.92 - 0.29En)$$

where En is the average enrichment of the assembly.

E. Uranium Mass Correction k4

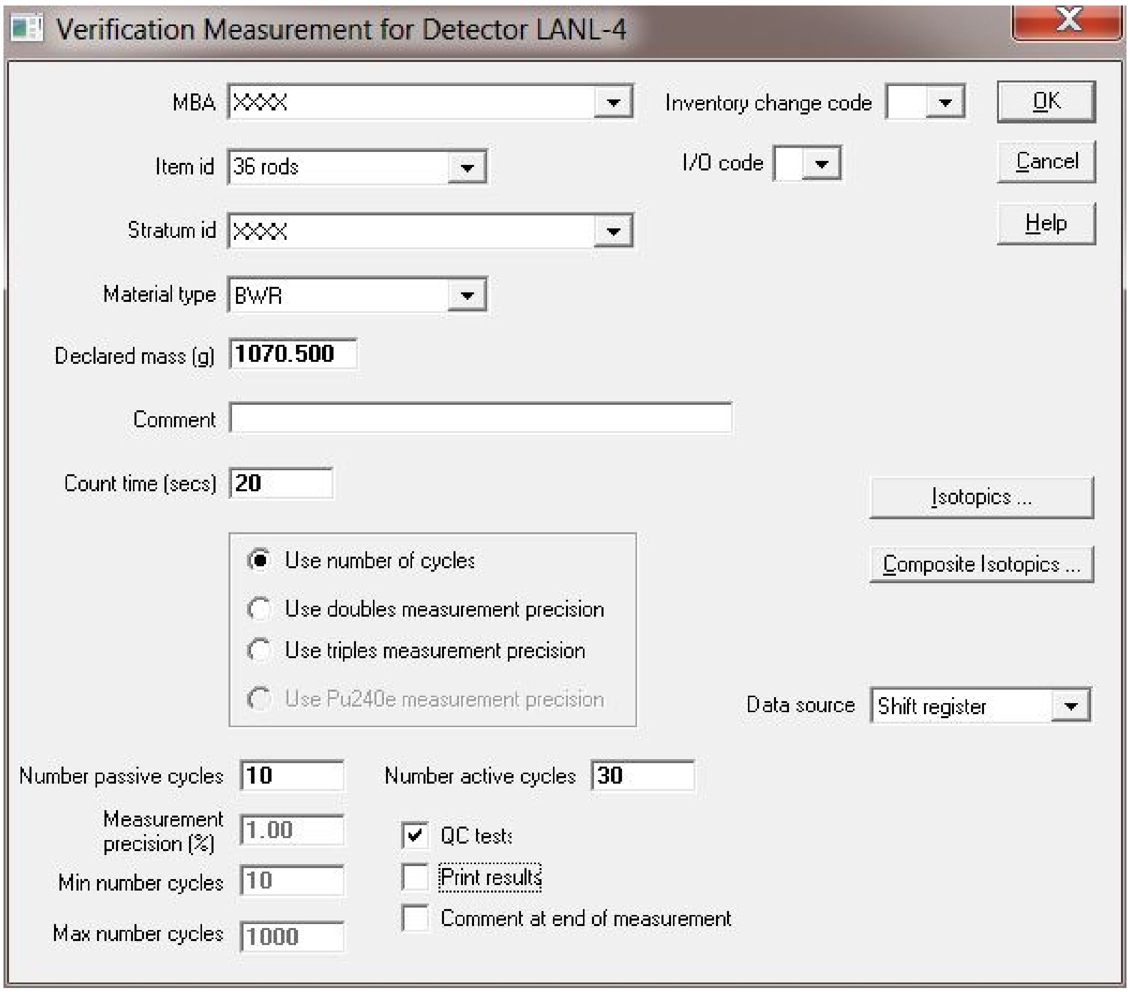
For some PWR assemblies the mass per unit length is very different from the calibration condition of 1215 g U/cm. In particular smaller PWR assemblies have loadings as small as ~900 g U/cm, thus lessening fast neutron multiplication, neutron scattering and end reflection from the extended fuel column. These reductions decrease the observed response. MCNP calculations and experiments were performed to establish a correction for the differences in the uranium loading between the standard and the unknowns. Our experience has shown that the k4 mass correction is not needed for BWR assemblies because the content of the current production loadings is similar (450-485 g U/cm). Our calibration reference assembly was 453 g U/cm and if the unknown assemblies are significantly different the correction for BWR is

$$k4 = 1 + 7.24 \times 10^{-4}(453 - U)$$

where U is the total uranium loading in g/cm. For the normal range of BWR fuel loadings this correction is only 1-2%. The same k4 correction is used for both the Cd and no Cd cases.

b. Verification Measurement

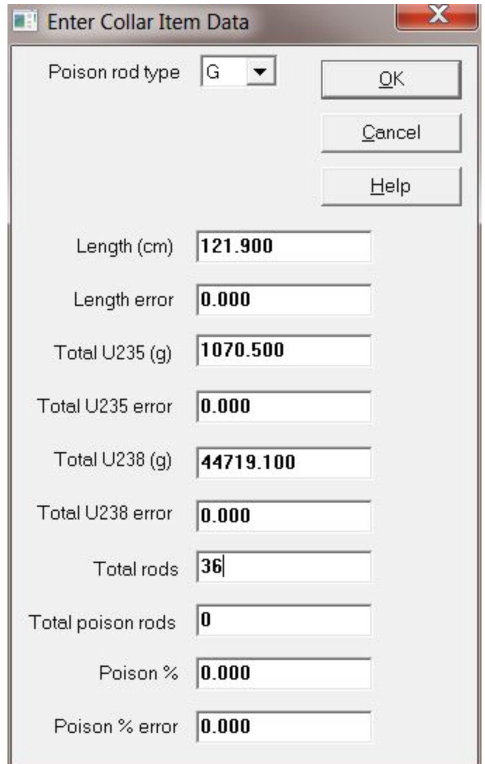
Place the collar around the fuel assembly. Select "Acquire | Verification" and enter the details of the assembly. The verification measurement menu will appear as shown below:

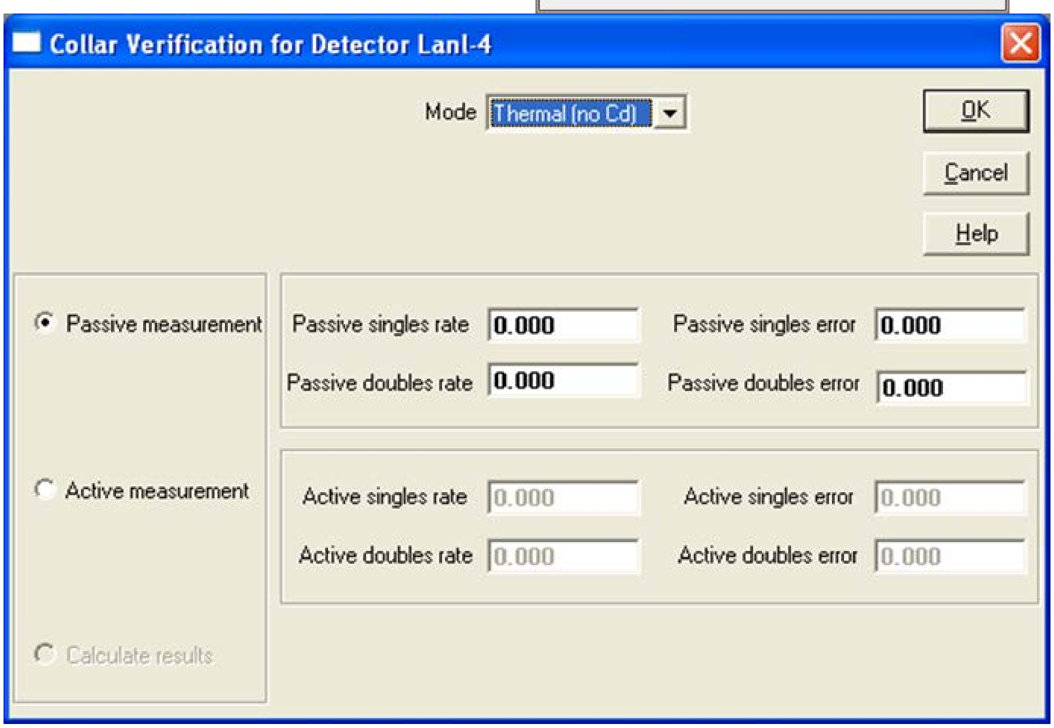


When you press OK, the next menu, shown to the right, allows you to enter more information about the collar. The ^{238}U value is the declared value for 36 rods. When all this information has been entered, press the OK button.

Next the additional correction factor screen will appear. Ensure that the steel rod box is marked. Press OK to confirm the k5 (steel rod) correction factor.

Next the main data acquisition control screen, shown below, will appear.





Under normal conditions you will select passive measurement and then OK. INCC will then start to collect the passive data. When it is finished, INCC will fill in the passive data fields. **AFTER** the instructor has put the AmLi source in the collar, select Active Measurement radio button and press OK. INCC will then take the active data. When it has finished you will be able to select Calculate results radio button to get the measured values.

The printout will contain the net passive Doubles rate (from ^{238}U) and the net active Doubles rate (from induced fission in ^{235}U caused by AmLi neutrons. The printout will also show the Net Corrected Doubles rate after all the correction factors are applied. This corrected Doubles rate is used to calculate the ^{235}U g/cm from the calibration curve which will then be multiplied by the active length to get the total mass of the assembly. Your result should be within 3 standard deviations of the declared value. Fill in the answers in the table below.

Declared Mass (g ^{235}U)	Corrected Doubles (cnts/s)	Measured Mass (g ^{235}U)	Operator Inspector Difference (OID) %	Uncertainty on OID %